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(54) **Adhesive tape and substrate for adhesive tape**

(57) The present invention provides an adhesive tape containing a substrate and an adhesive layer formed on at least one side of the substrate, wherein the substrate contains an olefin polymer and a flame retardant, but substantially no halogen atom, and the adhesive tape has a thermal deformation at 100°C of not more than 65%. The present invention also provides a substrate containing an olefin polymer and a flame retardant, but substantially no halogen atom, wherein the olefin polymer contains the following Component A and Component B:

Component A: a thermoplastic resin having a carbonyl oxygen atom in the molecular skeleton

Component B: a polymer alloy containing an ethylene component and a propylene component.

The adhesive tape and the substrate for the adhesive tape of the present invention are free of generation of dioxin and toxic gas upon incineration, and show superior resistance to thermal deformation, flame resistance and superior stretchability in combination.

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## Description

**[0001]** The present invention relates to an adhesive tape having a superior flame resistance and a substrate for the adhesive tape.

**[0002]** A conventional adhesive tape comprising a substrate for an adhesive tape (hereinafter to be also referred to simply as a substrate), which is made from polyvinyl chloride (hereinafter to be abbreviated as PVC), is superior in mechanical properties (particularly flexibility and stretchability), flame resistance, resistance to thermal deformation, and electrical insulation property. Combined with its beneficial economical aspect, the above-mentioned tape has been widely used as an insulating tape for electric equipment used in the fields of vehicles, such as automobiles, trains, buses, aircrafts and ships, as well as in houses and plants.

**[0003]** Particularly, an adhesive tape to be wound around a wire harness to be used for electric wires in e.g. automobiles, a coil for household electric appliances and electric wires is required to show high flame resistance (oxygen index of not less than 25%) and high resistance to thermal deformation. To meet these requirements, an adhesive tape using PVC as a substrate has been widely used.

**[0004]** Given the recent rise of consciousness toward environmental problems, however, there is a movement to reduce use of PVC and replace it with a material causing less environmental burden. This is because PVC is suspected of generating dioxin and toxic gas, such as chlorine gas, upon incineration. To replace PVC, the use of a polyolefin resin as a substrate has been considered, because it less likely generates dioxin or toxic gas, such as chlorine gas, upon incineration. However, polyolefin resins easily burn as compared to PVC and require addition of a flame retardant. For example, US-A-4,067,847 proposes addition of an inorganic flame retardant, such as magnesium hydroxide and aluminum hydroxide, which places only a small burden on the environment.

**[0005]** In consideration of the flexibility and stretchability necessary for an adhesive tape, e.g. polypropylene, ethylene-propylene copolymer and high density polyethylene having a relatively high melting point, are not suitable as the polyolefin resin used along with a flame retardant. Instead, a polyolefin resin having a low melting point needs to be used, such as low density polyethylene, linear low density polyethylene, ultra-low density polyethylene, middle density polyethylene, ethylene-vinyl acetate copolymer or ethylene-ethyl acrylate copolymer. These polyolefin resins having a low melting point are flexible and stretch easily, but are susceptible to thermal deformation.

**[0006]** As a method for improving resistance to thermal deformation of a resin film, there have been known a method comprising exposing the film to ionizing radiation, a method comprising previous addition of a crosslinking agent, such as organic peroxide (e.g., dicumylperoxide), to a resin and vapor heating of a film after forming to provide a crosslinking structure in the film. These methods also increase the number of production steps and production cost to the level that makes their practical use unfeasible.

**[0007]** A different method comprises the use of a mechanically blended polymer mixture of polypropylene, ethylene-propylene copolymer or high density polyethylene, having a relatively high melting point, and an elastomer consisting of ethylene copolymer, such as EPM (ethylene-propylene copolymer rubber), EBR (ethylene-butene rubber) and EPDM (ethylene-propylene-diene copolymer rubber). When a flame retardant is added to this polymer mixture, a molded product thereof becomes very stiff and lacks flexibility at room temperature, making stretchability strikingly low and resistance to thermal deformation insufficient.

**[0008]** It is therefore an object of the present invention to provide an adhesive tape free of generation of dioxin and toxic gas upon incineration, which has high levels of resistance to thermal deformation and flame resistance, as well as a substrate to be used for this adhesive tape.

**[0009]** It is also an object of the present invention to provide an adhesive tape having a high level of stretchability that is beneficial in winding a tape, in addition to high levels of the resistance to thermal deformation and flame resistance, as well as a substrate to be used for this adhesive tape.

**[0010]** According to the present invention, a thermoplastic resin having a carbonyl oxygen atom in the molecular skeleton and a polymer alloy containing an ethylene component and a propylene component, in combination, are used as an olefin polymer, and a flame retardant is added to give a substrate. As a result, a substrate for an adhesive tape, which shows high levels of resistance to thermal deformation and stretchability, as well as a high level of flame resistance, can be obtained.

**[0011]** Accordingly, the present invention provides an adhesive tape comprising a substrate and an adhesive layer formed on at least one side of the substrate, wherein the substrate comprises an olefin polymer and a flame retardant, but substantially no halogen atom, and the adhesive tape shows a thermal deformation at 100°C of not more than 65%.

**[0012]** In a preferable embodiment, the adhesive tape shows the following properties. It shows an elongation at break of not less than 150% at a tension speed of 300 mm/min. It shows a dynamic storage modulus (E') at 80°C of not less than 25 MPa and a dynamic storage modulus (E') at 120°C of not less than 10 MPa.

**[0013]** The present invention provides a substrate for an adhesive tape, which contains an olefin polymer and a flame retardant but contains substantially no halogen atom, wherein the olefin polymer contains the following Component A and Component B:

Component A: a thermoplastic resin having a carbonyl oxygen atom in the molecular skeleton

Component B: a polymer alloy containing an ethylene component and a propylene component

[0014] In a preferable embodiment, the Component B shows the following properties. It shows a dynamic storage modulus (E') at 80°C of not less than 40 MPa and less than 180 MPa, and a dynamic storage modulus (E') at 120°C of not less than 12 MPa and less than 70 MPa. It shows a dynamic storage modulus (E') at 23°C of not less than 200 MPa and less than 400 MPa.

[0015] Component A is an ethylene copolymer having a melting point of not more than 120°C, which is obtained by polymerizing a vinyl ester compound and/or  $\alpha,\beta$ -unsaturated carboxylic acid or a derivative thereof, or a metal salt of the ethylene copolymer. Component A and Component B are mixed at a weight ratio (A:B) of 1:9 - 8:2. The flame retardant is added in an amount of 20 - 200 parts by weight per 100 parts by weight of an olefin polymer. The flame retardant is a metal hydroxide.

[0016] In another preferable embodiment, the substrate has a dynamic storage modulus (E') at 80°C of not less than 25 MPa, and a dynamic storage modulus (E') at 120°C of not less than 10 MPa. The substrate is not crosslinked during or after its formation.

[0017] Fig. 1(a) and Fig. 1(b) show a method for the evaluation of thermal deformation of an adhesive tape, wherein Fig. 1(a) is a side view of the test sample, Fig. 1(b) is a side view of a test apparatus, 1 is a round bar, 2 is a loading plate, 2a is a convex, 3 is a parallel plate, 10 is a test sample and T is an adhesive tape.

[0018] The adhesive tape of the present invention comprises a substrate containing an olefin polymer and a flame retardant, but contains substantially no halogen atom, and an adhesive layer formed at least on one side of the substrate. The adhesive tape has a thermal deformation at 100°C of not more than 65% (preferably not more than 40%, particularly preferably 0%), and preferably the thermal deformation of not more than 65% together with an elongation at break of not less than 150% (preferably 170-500%) at a tension speed of 300 mm/min.

[0019] As used herein, by the "substantially no halogen atom" is meant non-use of a substance having a halogen atom in a molecule as a material of the substrate. However, a halogen atom detected in an ultra-trace amount by analysis of the composition of a tape substrate using mechanical analysis means is excluded. For example, a halogen atom may be contained in an ultra-trace amount in the substrate, due to the use of a halogen-containing substance as a catalyst for the synthesis of a constituting material.

[0020] A thermal deformation at 100°C and an elongation at break (%) at a tension speed of 300 mm/min of the adhesive tape are respectively used as indices of resistance to thermal deformation and stretchability of the tape. An adhesive tape having a thermal deformation at 100°C of not more than 65% shows a high level of resistance to thermal deformation in actual use (adhered to a target object). An adhesive tape having an elongation at break of not less than 150% at a tension speed of 300 mm/min shows a high level of stretchability. Therefore, the tape can be easily stretched in actual use (adhered to a target object (winding)) and the adhesion (winding) can be performed with superior workability. The thermal deformation and the elongation at break of the adhesive tape were measured by the following method.

#### Measurement method of thermal deformation

[0021] Measured according to UL-510. As shown in Figure 1(a), an adhesive tape T is wound around a round conductor bar 1 having a diameter (d) of 2 mm and a length of 30 mm to prepare a test sample 10. The outer diameter (D0) of the test sample 10 at normal temperature is measured with a dial gauge defined in JIS B 7503, vernier calipers defined in JIS B 7507, or a measurement tool having the same degree of measurement precision. The test sample 10 is placed in a tester heated to a predetermined temperature ( $100^{\circ}\text{C} \pm 1.0^{\circ}\text{C}$ ) and heated for 60 minutes. The test sample is placed between a loading plate 2 having a columnar convex 2a having a diameter of  $9.5 \pm 0.2$  mm and a parallel plate 3, as shown in Figure 1(b), pressurized (4.9 N) from the vertical direction of the planar surface of the plates and left standing at a predetermined temperature ( $100^{\circ}\text{C} \pm 1.0^{\circ}\text{C}$ ) for 60 minutes. The test sample is measured for the outer diameter (D1) *in situ*. The thickness (t0) before heating the tape and the thickness (t1) after heating the tape are calculated from the following formula (II). A decrease in the thickness (thermal deformation (X)) of the tape is calculated from the following formula (III):

$$t = (D - d) / 2 \quad (\text{II})$$

[0022] wherein D is an outer diameter of the test sample and d is a diameter of the round bar,

$$X(\%) = \{(t_0 - t_1) / t_0\} \times 100 \quad (\text{III})$$

wherein t0 is a thickness (mm) before heating and t1 is a thickness (mm) after heating.

Measurement method of elongation at break

**[0023]** A test piece (width 19 mm, length 100 mm) taken from an adhesive tape prepared by forming a 30  $\mu$ m thick adhesive layer on one side of a 0.2 mm thick substrate is held at both ends in the length direction thereof at 23°C, 60% RH, wherein the length direction is the direction of flow during forming a substrate (MD direction), and stretched at a tension speed of 300 mm/min in the length direction by a universal tensile compression tester to measure elongation at break (%).

**[0024]** In the present invention, an adhesive tape having a thermal deformation at 100°C of not more than 65% can be obtained by using Component A and Component B to be mentioned later as olefin polymers together with a flame retardant, by appropriately determining the kind, mixing weight ratio and combination of Component A and Component B, by using Component B having a specific dynamic storage modulus ( $E'$ ) and/or by using Component A having a specific melting point.

**[0025]** This also applies to an adhesive tape having a thermal deformation of not more than 65% and an elongation at break of not less than 150% at a tension speed of 300 mm/min.

**[0026]** In the present invention, the olefin polymer constituting a tape substrate can be used without any particular limitation as long as a mixture of the olefin polymer and a flame retardant in an amount to bring about sufficient flame resistance can impart desired superior resistance to thermal deformation and stretchability to an adhesive tape. An olefin polymer mainly containing the following Component A and Component B is preferable.

Component A: a thermoplastic resin having a carbonyl oxygen atom in the molecular skeleton

Component B: a polymer alloy containing an ethylene component and a propylene component

**[0027]** The thermoplastic resin having a carbonyl oxygen atom (oxygen atom attributable to carbonyl) in the molecular skeleton of Component A activates a flame resistance-imparting action by a flame retardant and imparts appropriate flexibility and fine stretchability to a substrate (adhesive tape). Particularly, a soft polyolefin resin having a carbonyl oxygen atom in the molecular skeleton is preferable. The soft polyolefin resin having a carbonyl oxygen atom in the molecular skeleton may be an ethylene copolymer or a metal salt thereof (particularly ionomer) comprising a vinyl ester compound and/or an  $\alpha,\beta$ -unsaturated carboxylic acid or a derivative thereof as a monomer or a comonomer. In general, it has a melting point of not more than 120°C, preferably 40-100°C. The melting point is measured by a differential scanning calorimeter (DSC).

**[0028]** In the above-mentioned ethylene copolymer or a metal salt thereof (particularly ionomer), the vinyl ester compound may be, for example, a lower alkyl ester of vinyl alcohol such as vinyl acetate. The  $\alpha,\beta$ -unsaturated carboxylic acid or a derivative thereof may be, for example, unsaturated carboxylic acids and anhydrides thereof, such as acrylic acid, methacrylic acid, maleic acid, fumaric acid, maleic anhydride and itaconic anhydride; and unsaturated carboxylates such as methyl acrylate, methyl methacrylate, ethyl acrylate, ethyl methacrylate, propyl acrylate, propyl methacrylate, isopropyl acrylate, isopropyl methacrylate, n-butyl acrylate, n-butyl methacrylate, cyclohexyl acrylate, cyclohexyl methacrylate, stearyl acrylate, stearyl methacrylate, lauryl acrylate, lauryl methacrylate, monomethyl maleate, monoethyl maleate, diethyl maleate, monomethyl fumarate, glycidyl acrylate and glycidyl methacrylate. Of these, an alkyl (meth)acrylate, particularly ethyl acrylate, is preferable.

**[0029]** Examples of suitable ethylene copolymer or a metal salt thereof (particularly ionomer) include ethylene-acrylic acid copolymer, ethylene-methacrylic acid copolymer, ethylene-ethyl acrylate copolymer, ethylene-acrylic acid-ethyl acrylate copolymer, ethylene-vinyl acetate-copolymer, ethylene-vinyl acetate-ethyl acrylate copolymer, ethylene-glycidyl methacrylate copolymer, ethylene-glycidyl methacrylate-ethyl acrylate copolymer and a metal salt thereof (particularly ionomer) which may be used alone or in combination.

**[0030]** The effect of activating a flame resistance-imparting action of a flame retardant by the thermoplastic resin having a carbonyl oxygen atom in the molecular skeleton of Component A cannot be achieved by a thermoplastic resin without a carbonyl oxygen atom in the molecular skeleton.

**[0031]** The polymer alloy containing an ethylene component and a propylene component of Component B mainly suppresses thermal deformation of a substrate (adhesive tape). By making an alloy of an ethylene component and a propylene component, the polymer alloy shows viscoelasticity suitable for suppressing thermal deformation of particularly a substrate (adhesive tape).

**[0032]** The constitution (form) of the polymer alloy is not particularly limited. For example, various constitutions (forms) are acceptable such as (1) a polymer blend (physical mixture) of two or more kinds of polymers physically mixed, (2) a block copolymer or graft copolymer wherein two or more kinds of polymers are bonded by a covalent bond, and (3) an IPN (Interpenetrating Polymer Network) structure wherein two or more kinds of polymers are intertwined without a covalent bond.

**[0033]** The polymer alloy may not be homogeneous in composition but may have a distribution. In addition, it may be an alloy of two or more kinds of compatible polymers or a phase separation structure formed by two or more kinds

of incompatible polymers. It may have a thermal property showing plural exothermic or endothermic peaks by DSC measurement.

**[0034]** Examples of the polymer alloy containing an ethylene component and a propylene component of Component B include a physical mixture of polypropylene (homo polypropylene, random polypropylene) and polyethylene (inclusive of copolymer of ethylene and a small amount of other  $\alpha$ -olefin), propylene/ethylene copolymer (block copolymer or graft copolymer), terpolymer of propylene, ethylene and  $\alpha$ -olefin other than these. As used herein, by the above-mentioned other  $\alpha$ -olefin is meant e.g. 1-butene, 1-penten, 1-hexene, 4-methyl-1-pentene, 1-heptene and 1-octene with preference given to 1-butene. By the homo polypropylene is meant a polymer comprising substantially 100% of propylene and by the random polypropylene is meant a copolymer comprising several percent of ethylene randomly copolymerized.

**[0035]** When the polymer alloy is a copolymer, a copolymer obtained by multi-step polymerization involving two or more steps is preferable, with preference given to a propylene/ethylene copolymer. The copolymer obtained by multi-step polymerization involving two or more steps can be obtained by, as disclosed in JP-A-4-224809, for example, preliminarily polymerizing propylene or propylene and  $\alpha$ -olefin other than propylene in the presence of a titanium compound and an organic aluminum compound as catalysts in the first step of the multi-step polymerization, and then copolymerizing propylene and ethylene (after addition of  $\alpha$ -olefin other than propylene and ethylene as necessary) in the presence of the produced titanium-containing polyolefin and an organic aluminum compound in the second step or a subsequent step. As a result, the obtained is a copolymer of a polymer produced in the first step [e.g., polypropylene (homopolymer of propylene) or propylene- $\alpha$ -olefin copolymer] and a polymer produced in the second step or a subsequent step (e.g., propylene-ethylene copolymer or propylene-ethylene-other  $\alpha$ -olefin copolymer), which copolymer has been blended to a molecular level in the second step or a subsequent step. The above-mentioned titanium compound is, for example, a solid catalyst having an average particle size of 15  $\mu$ m and is obtained by pulverizing titanium trichloride and magnesium chloride together, and treating with e.g. n-butyl orthotitanate, 2-ethylhexanol, ethyl p-toluate, silicon tetrachloride or diisobutyl phthalate. The organic aluminum compound may be, for example, alkyl aluminum such as triethyl aluminum. In a polymerized phase, a silicon compound such as diphenyldimethoxysilane or an iodine compound such as ethyl iodide may be contained as an electron donor.

**[0036]** The polymer alloy containing an ethylene component and a propylene component of Component B preferably shows high elasticity under high temperature, that is, in consideration of the temperature during use of the adhesive tape, it preferably shows a dynamic storage modulus ( $E'$ ) at 80°C of not less than 40 MPa and less than 180 MPa (preferably within the range of 45 - 160 MPa) and a dynamic storage modulus ( $E'$ ) at 120°C of not less than 12 MPa and less than 70 MPa (preferably within the range of preferably 15 - 65 MPa). The above-mentioned dynamic storage modulus ( $E'$ ) sufficiently suppresses thermal deformation of a substrate (adhesive tape).

**[0037]** The dynamic storage modulus ( $E'$ ) here is measured in the following manner. That is, a polymer alloy test piece (thickness 0.2 mm: width 10 mm, length 20 mm) is prepared, and the dynamic viscoelastic behavior of this test piece at dispersed temperatures is measured with DMS200 (trademark, manufactured by Seiko Instruments) as a measurement apparatus under the measurement conditions of method: extension mode, temperature rise: 2°C/min, frequency: 1 Hz.

**[0038]** Examples of the polymer alloy showing such dynamic storage modulus ( $E'$ ) include trademarks CATALLOY KS-353P, CATALLOY KS-021P and CATALLOY C200F manufactured by Montell SKD Sunrise Ltd., and the like.

**[0039]** The polymer alloy of Component B preferably shows a dynamic storage modulus ( $E'$ ) at 23°C of not less than 200 MPa and less than 400 MPa in consideration of the workability of the adhesive tape at near room temperature (followability of the adhesive tape to the object to be adhered to). The dynamic storage modulus ( $E'$ ) of this range of the polymer alloy of Component B leads to fine flexibility of the substrate, and therefore, improved followability to the object to be adhered to. The dynamic storage modulus ( $E'$ ) here is measured by the aforementioned method. Examples of the polymer alloy having such dynamic storage modulus ( $E'$ ) include trademarks CATALLOY KS-353P, CATALLOY KS-021P and CATALLOY C200F manufactured by Montell SKD Sunrise Ltd.

**[0040]** Component A (a thermoplastic resin having a carbonyl oxygen atom in the molecular skeleton) and Component B (a polymer alloy containing an ethylene component and a propylene component) are mixed at a weight ratio (A:B) of generally 1:9 - 8:2, preferably 2:8 - 6:4. When the weight ratio is outside this range and Component A is used in a smaller amount (Component B is in a greater amount), the substrate may be poor in flexibility, stretchability and flame resistance. When Component A is used in a greater amount (Component B is in a smaller amount), the substrate may show poor resistance to thermal deformation.

**[0041]** In the present invention, the flame retardant may be, for example, a metal hydroxide such as aluminum hydroxide, magnesium hydroxide, zirconium hydroxide, calcium hydroxide and barium hydroxide; metal carbonate such as basic magnesium carbonate, magnesium calcium carbonate, calcium carbonate, barium carbonate and dolomite; metal hydrate (hydrate of metal compound) such as hydrotalcite and borax; barium metaborate or magnesium oxide. These may be used alone or in combination. Of these, one selected from the group of metal hydroxides such as aluminum hydroxide, magnesium hydroxide, zirconium hydroxide, calcium hydroxide and barium hydroxide, basic magne-

sium carbonate and hydrotalcite, particularly the above-mentioned metal hydroxide, is advantageous in view of flame resistance-imparting effect and economical aspect.

**[0042]** The flame retardant has a particle size that varies depending on the kind of compound. For example, a metal hydroxide, such as aluminum hydroxide, magnesium hydroxide and the like, has an average particle size of about 0.1 - 50  $\mu\text{m}$ , preferably 0.5 - 20  $\mu\text{m}$ . The particle size here is measured by a laser diffraction method.

**[0043]** The flame retardant is added in an amount of generally 20 - 200 parts by weight, preferably 40 - 150 parts by weight, per 100 parts by weight of the olefin polymer. When the amount added of the flame retardant is outside this range and is smaller, sufficient flame resistance cannot be imparted to the substrate (adhesive tape). When it is greater, the substrate (adhesive tape) may become poor in flexibility and stretchability.

**[0044]** With the purpose of encouraging charring of a flame retardant to give a carbonized layer, red phosphorus may be used. When using red phosphorus, to prevent generation of toxic phosphine even when red phosphorus is heated in the presence of water (stabilization of surface of red phosphorus), red phosphorus may be coated with a film made from metal hydroxide selected from aluminum hydroxide, magnesium hydroxide, zinc hydroxide and titanium hydroxide. Alternatively, a film of a thermosetting resin (e.g. phenol resin) may be formed on the metal hydroxide film to cover red phosphorus in duplicate. A char forming auxiliary containing such red phosphorus is suitably added in a proportion of generally 2 - 10 parts by weight, preferably 4 - 8 parts by weight, per 100 parts by weight of the olefin polymer.

**[0045]** When a char forming auxiliary containing red phosphorus is used, at least one member selected from carbon black, borates and silicone compounds (e.g. silicone oil, silicone rubber or silicone resin) is concurrently used to secure a more preferable effect. In this case, at least one member selected from carbon black, borates and silicone compounds is added in an amount of generally 0.5 - 10 wt%, preferably 1 - 5 wt%, per 100 parts by weight of the olefin polymer.

**[0046]** In the present invention, the substrate consists of an olefin polymer and a flame retardant as essential components. Where necessary, an inorganic filler such as titanium oxide and zinc oxide, an age resister and antioxidant such as amine type, quinoline type, hydroquinone type, phenol type, phosphorous type and phosphite type age resisters, an ultraviolet absorber such as salicylic acid derivative, benzophenone type, benzotriazole type and hindered amine type ultraviolet absorbers, a lubricant and a plasticizer may be added.

**[0047]** According to the present invention, the substrate is generally obtained by dry blending a polyolefin polymer, a flame retardant and a substance added as necessary such as filler and the like, kneading the mixture in a Banbury mixer, a roller or an extruder, and forming the kneaded mixture into a film by a known forming method such as compression forming, calendar forming, injection forming or extrusion forming.

**[0048]** The substrate (film) has a thickness of generally 0.01 - 1 mm, preferably 0.05 - 0.5 mm, though subject to change depending on the use of the adhesive tape.

**[0049]** After film forming, the substrate of the present invention preferably does not undergo a crosslinking treatment comprising an ionizing radiation such as electron beam,  $\beta$ -ray and  $\gamma$ -ray, after film forming, or a crosslinking treatment comprising addition, during forming process, of a crosslinking agent, such as organic peroxide, or a crosslinking auxiliary to the material to be formed. In other words, the whole substrate is preferably free of crosslinking structure.

**[0050]** The adhesive tape of the present invention is produced by forming an adhesive layer at least on one side of the above-mentioned substrate. Examples of the adhesive include any adhesive, such as rubber type, hot melt type, acrylic type and emulsion type adhesives, which is currently in use. Examples of preferable base polymer of the rubber and hot melt adhesives include natural rubber, regenerated rubber, silicone rubber, isoprene rubber, styrene-butadiene rubber, polyisoprene, NBR, styrene-isoprene copolymer and styrene-isoprene-butadiene copolymer.

**[0051]** Examples of the tackifier to be used for adhesive include rosin tackifier, terpene tackifier, aliphatic petroleum hydrocarbon (C5) tackifier, aliphatic petroleum hydrocarbon (C9) tackifier and hydrogenated compounds. It is also possible to add an additive usually added to the adhesive for an adhesive tape, such as oil, wax and antioxidant in an amount determined by a conventional method.

**[0052]** Of the aforementioned adhesives, an acrylic adhesive is preferable. Examples thereof include homopolymer of (meth)acrylate or a copolymer thereof with a copolymerizable monomer. Examples of (meth)acrylate and the copolymerizable monomer include alkyl (meth)acrylate (e.g., methyl ester, ethyl ester, butyl ester, 2-ethylhexyl ester and octyl ester), glycidyl (meth)acrylate, (meth)acrylic acid, itaconic acid, maleic anhydride, (meth)acrylamide, (meth)acrylic N-hydroxyamide, alkylaminoalkyl (meth) acrylate (e.g., dimethylaminoethyl methacrylate and t-butylaminoethyl methacrylate), vinyl acetate, styrene and acrylonitrile.

**[0053]** Of these, the main monomer is preferably alkyl acrylate wherein its homopolymer (single polymer) generally has a glass transition temperature of not more than  $-50^{\circ}\text{C}$ .

**[0054]** The adhesive is applied by a conventionally-known method such as cast method, roll coater method, reverse coater method or doctor blade method.

**[0055]** The adhesive layer has a thickness (thickness after drying) that varies depending on the use of the adhesive tape, which is generally 10 - 50  $\mu\text{m}$ , preferably 15 - 40  $\mu\text{m}$ .

**[0056]** In the present invention, the adhesive tape preferably has a dynamic storage modulus ( $E'$ ) at  $80^{\circ}\text{C}$  of not less than 25 MPa and a dynamic storage modulus ( $E'$ ) at  $120^{\circ}\text{C}$  of not less than 10 MPa. Such dynamic storage moduli

make the substrate (adhesive tape) less likely suffer from thermal deformation. In view of the suitable flexibility and stretchability necessary for an adhesive tape, the dynamic storage modulus (E') at 80°C is preferably not more than 200 MPa, and the dynamic storage modulus (E') at 120°C is preferably not more than 150 MPa.

[0057] The dynamic storage modulus (E') here of the adhesive tape is measured in the following manner. That is, a test piece (width 10 mm, length 20 mm) is taken from an adhesive tape comprising a substrate (thickness 0.2 mm) and an adhesive layer formed thereon, and the dynamic viscoelastic behavior of this test piece at dispersed temperatures is measured with DMS200 (trademark, manufactured by Seiko Instruments) as a measurement apparatus under the measurement conditions of method: extension mode, temperature rise: 2°C/min, frequency: 1 Hz.

[0058] The dynamic storage modulus (E') of the adhesive tape is free from an effect of the adhesive layer. That is, the presence of an adhesive layer does not change the value of the dynamic storage modulus (E'), and the dynamic storage modulus (E') is substantially that of the substrate.

[0059] In the present invention, an adhesive tape (substrate) having "a dynamic storage modulus (E') at 80°C of not less than 25 MPa and a dynamic storage modulus (E') at 120°C of not less than 10 MPa" can be obtained by using the above-mentioned Component A and Component B as olefin polymers together with a flame retardant, by appropriately determining the kind, mixing weight ratio and/or combination of Component A and Component B, by using Component B having a specific dynamic storage modulus (E') and/or by using Component A having a specific melting point.

[0060] The present invention is explained in detail in the following by referring to Examples and Comparative Examples. The present invention is not limited by these examples.

#### Materials used

Component A (thermoplastic resin having a carbonyl oxygen atom in the molecular skeleton)

#### [0061]

A1: ethylene-vinyl acetate copolymer (EVA), melting point 84°C (trademark: EVAFLEX P-1905, Du Pont-Mitsui Polychemicals Company, Ltd.)

A2: ethylene-ethyl acrylate copolymer (EEA), melting point 84°C (trademark: EVAFLEX A-702, Du Pont-Mitsui Polychemicals Company, Ltd.)

Component B (polymer alloy containing an ethylene component and a propylene component)

#### [0062]

B1: CATALLOY KS-353P (trademark, Montell SKD Sunrise Ltd.)

B2: CATALLOY KS-021P (trademark, Montell SKD Sunrise Ltd.)

B3: CATALLOY C200F (trademark, Montell SKD Sunrise Ltd.)

[0063] The dynamic storage moduli (E') at 23°C, 80°C and 120°C of the above-mentioned B1 - B3 are as follows:

B1 (23°C: 210 MPa, 80°C: 52 MPa, 120°C: 21 MPa)

B2 (23°C: 294 MPa, 80°C: 125 MPa, 120°C: 59 MPa)

B3 (23°C: 303 MPa, 80°C: 65 MPa, 120°C: 20 MPa)

Component C (flame retardant)

#### [0064]

C1: magnesium hydroxide (Mg(OH)<sub>2</sub>) (trademark: Magshizu N-3, Konoshima Chemical Industrial Co., Ltd.)

C2-1: red phosphorus (trademark: Hishigard CP-A15, The Nippon Chemical Industrial Co., Ltd.)

C2-2: red phosphorus (trademark: Norbaexcell F5, Phosphorus Chemical Industry Co., Ltd.)

C3: carbon black (trademark: Shisuto 3H, TOKAI CARBON CO., LTD.)

#### Preparation of substrate and adhesive tape

[0065] The above-mentioned Component A, Component B and Component C were dry blended, kneaded in a pressurized 3L kneader at 180°C and pelleted. The composition was formed into a 0.2 mm thick film by a T die extruder to give a substrate. One side surface of the substrate was subjected to a corona discharge treatment and an acrylic

adhesive prepared by the following method was applied (thickness 30  $\mu\text{m}$ ) to give an adhesive tape.

Preparation of acrylic adhesive

[0066] In a reaction vessel equipped with a condenser, a nitrogen inlet, a thermometer and a stirrer were placed, in a toluene solvent, 2-ethylhexyl acrylate (100 parts by weight), acrylic acid (2 parts by weight), and benzoyl peroxide (0.2 part by weight) as a polymerization initiator, and the mixture was allowed to react at 60°C for 8 hours to give a polymer. To this polymer solution was added a polyisocyanate crosslinking agent (trademark: CORONATE L, NIPPON POLY-URETHANE INDUSTRY Co., LTD., 3 parts by weight per 100 parts by weight of the polymer solid content) to give an acrylic adhesive.

**Examples 1 - 7**

[0067] From Component A, Component B and Component C of the materials and amounts shown in the following Table 1, a substrate (adhesive tape) was prepared according to the above-mentioned forming method.

**Comparative Example 1**

[0068] Using Component A (A1: 100 parts by weight) but without using Component B or Component C, a substrate (adhesive tape) was prepared according to the above-mentioned forming method.

**Comparative Example 2**

[0069] Using Component A (A2: 100 parts by weight) but without using Component B or Component C, a substrate (adhesive tape) was prepared according to the above-mentioned forming method.

**Comparative Example 3**

[0070] Using Component B (B1: 100 parts by weight) but without using Component A or Component C, a substrate (adhesive tape) was prepared according to the above-mentioned forming method.

**Comparative Example 4**

[0071] Using polypropylene (PP) (100 parts by weight, trademark: NOVATEC FX3, Japan Polychem Corporation) but without using Component A, Component B or Component C, a substrate (adhesive tape) was prepared according to the above-mentioned forming method.

**Comparative Example 5**

[0072] Using Component A (A1: 100 parts by weight) and Component C (C1: 50 parts by weight) but without using Component B, a substrate (adhesive tape) was prepared according to the above-mentioned forming method.

**Comparative Example 6**

[0073] Using Component A (A1: 100 parts by weight) and Component C (C1: 100 parts by weight) but without using Component B, a substrate (adhesive tape) was prepared according to the above-mentioned forming method.

**Comparative Example 7**

[0074] Using Component A (A1: 100 parts by weight) and Component C (C1: 105 parts by weight, C2-1: 5 parts by weight) but without using Component B, a substrate (adhesive tape) was prepared according to the above-mentioned forming method.

**Comparative Example 8**

[0075] Using Component A (A1: 100 parts by weight) and Component C (C1: 50 parts by weight, C2-2: 4 parts by weight, C3: 2 parts by weight) but without using Component B, a substrate (adhesive tape) was prepared according to the above-mentioned forming method.



Evaluation Test

**[0076]** The substrates (adhesive tapes) of Examples 1 - 7 and Comparative Examples 1 - 8 were subjected to the following evaluation tests. The dynamic storage moduli (E') at 80°C and 120°C of the adhesive tape were measured by the aforementioned method.

Evaluation of flame resistance

**[0077]** At the step of forming into a 3 mm thick sheet using a press machine during the formation of the aforementioned substrate, a test piece (size: length 70 mm, width 6.5 mm) was taken from the sheet and this test piece was burnt according to the incineration test method of polymer material by the oxygen index method according to JIS K 7201 to examine if the test piece continued to burn for 3 minutes or longer. In addition, the amount of minimum oxygen flow necessary for continuous flame burning of not less than 50 mm thereof after flame burning, and the nitrogen flow at that time were measured by a flowmeter (Candle Type Flammability Tester, TOYO SEIKI SEISAKU-SHO Co., Ltd.). The oxygen index was calculated from the following formula (I) and the flame resistance was evaluated based on the oxygen index. The flame resistance of not less than 25% was an acceptance value and less than 25% was a rejection value.

$$\text{Oxygen index (O.I.)} = \{[O_2] / ([O_2] + [N_2])\} \times 100 \quad (I)$$

wherein  $[O_2]$  is an oxygen flow (l/min) and  $[N_2]$  is a nitrogen flow (l/min).

Evaluation of resistance to thermal deformation and stretchability

**[0078]** To evaluate resistance to thermal deformation, the thermal deformation of an adhesive tape was measured by the aforementioned method, wherein the thermal deformation of not more than 65% was an acceptance value. To evaluate stretchability, the elongation at break of the adhesive tape was measured by the aforementioned method, wherein the elongation at break of not less than 150% was an acceptance value.

**[0079]** These evaluation results and the dynamic storage moduli (E') at 80°C and 120°C of the adhesive tape are shown in the following Table 1.

Table 1

	Component A		Component B		Component C				Elonga- tion at break (%)	Thermal deforma- tion (%)	Dynamic storage modulus (E') (MPa)		Oxygen index index (%)
	Kind	Amount added	KInd	Amount added	C1 Amount added	C2		C3 Amount added					
						Kind	Amount added						
Example 1	A2	60	B1	40	100	C2-1	8	2	390	58	19	31	
Example 2	A1	50	B1	50	100	C2-2	8	2	310	45	67	32	
Example 3	A1	20	B1	80	100	C2-2	8	2	270	23	89	30	
Example 4	A1	20	B1	80	80	C2-2	8	2	470	25	84	28	
Example 5	A1	20	B2	80	100	C2-2	8	2	220	23	197	30	
Example 6	A1	20	B3	80	100	C2-2	8	2	230	23	148	29	
Example 7	A2	20	B1	80	50	C2-1	8	2	290	34	114	28	
Com. Ex. 1	A1	100	-	-	-	-	-	-	720	100	11	20	
Com. Ex. 2	A2	100	-	-	-	-	-	-	720	100	11	20	
Com. Ex. 3	-	-	B1	100	-	-	-	-	820	19	52	18	
Com. Ex. 4	-	-	PP	100	-	-	-	-	630	5	214	17	
Com. Ex. 5	A1	100	-	-	50	-	-	-	760	100	12	22	
Com. Ex. 6	A1	100	-	-	100	-	-	-	610	100	17	24	
Com. Ex. 7	A1	100	-	-	105	C2-1	5	-	280	100	16	38	
Com. Ex. 8	A1	100	-	-	50	C2-2	4	2	820	100	13	32	

[0080] In the Table, the amounts of Component A, Component B and Component C are in parts by weight, and ※ means that the adhesive tape (substrate) was melted and the dynamic storage modulus (E') could not be measured. The thermal deformation of 100% means that an adhesive tape was melted and dropped from the round bar, thus failing

to maintain the shape of a tape. The polypropylene (PP) used in Comparative Example 4 is conveniently listed in the column of Component B.

**[0081]** As is evident from the foregoing explanation, the substrate for an adhesive tape of the present invention contains an olefin polymer and a flame retardant but substantially no halogen atom, and the adhesive tape has a thermal deformation at 100°C of not more than 65% and an elongation at break of not less than 150% at a tension speed of 300 mm/min. Consequently, the adhesive tape has suitable flexibility, a high level of stretchability and a high level of resistance to thermal deformation, despite the existence of the flame retardant, and a high level of flame resistance, and is free of generation of dioxin and toxic gas, such as chlorine gas, upon incineration.

**[0082]** This adhesive tape can be sufficiently used as a substitute for an adhesive tape using a PVC substrate, which has been used as an insulating tape for electric equipment used in the fields of vehicles, such as automobiles, trains, buses, aircrafts and ships, as well as in houses and plants. Inasmuch as it is free of generation of dioxin and toxic gas upon incineration and places less burden on the environment, the adhesive tape is highly valuable for use.

### Claims

1. An adhesive tape comprising a substrate and an adhesive layer formed on at least one side of the substrate, wherein the substrate comprises an olefin polymer and a flame retardant, but substantially no halogen atom, and the adhesive tape has a thermal deformation at 100°C of not more than 65%.

2. The adhesive tape of claim 1, which has an elongation at break of not less than 150% at a tension speed of 300 mm/min.

3. The adhesive tape of claim 1 or claim 2, wherein the olefin polymer comprises the following Component A and Component B:

Component A: a thermoplastic resin having a carbonyl oxygen atom in the molecular skeleton

Component B: a polymer alloy containing an ethylene component and a propylene component.

4. The adhesive tape of claim 3, wherein the Component B has a dynamic storage modulus (E') at 80°C of not less than 40 MPa and less than 180 MPa, and a dynamic storage modulus (E') at 120°C of not less than 12 MPa and less than 70 MPa.

5. The adhesive tape of claim 3, wherein the Component B has a dynamic storage modulus (E') at 23°C of not less than 200 MPa and less than 400 MPa.

6. The adhesive tape of claim 3, wherein the Component A is an ethylene copolymer or a metal salt thereof, having a melting point of not more than 120°C, which is obtained by polymerizing a vinyl ester compound, or an  $\alpha,\beta$ -unsaturated carboxylic acid or a derivative thereof, or the vinyl ester compound and the  $\alpha,\beta$ -unsaturated carboxylic acid or a derivative thereof.

7. The adhesive tape of claim 3, wherein the Component A and the Component B are mixed at a weight ratio (A:B) of 1:9 - 8:2.

8. The adhesive tape of claim 1 or claim 2, wherein the flame retardant is added in an amount of 20 - 200 parts by weight per 100 parts by weight of the olefin polymer.

9. The adhesive tape of claim 1 or claim 2, wherein the flame retardant is a metal hydroxide.

10. The adhesive tape of claim 1 or claim 2, which has a dynamic storage modulus (E') at 80°C of not less than 25 MPa and a dynamic storage modulus (E') at 120°C of not less than 10 MPa.

11. The adhesive tape of claim 1 or claim 2, wherein the substrate is not crosslinked during or after a forming process thereof.

12. A substrate for an adhesive tape, which comprises an olefin polymer and a flame retardant, but substantially no halogen atom, wherein the olefin polymer and the flame retardant are defined as in any one of claims 3 to 9.

13. The substrate of claim 12, which has a dynamic storage modulus (E') at 80°C of not less than 25 MPa and a

dynamic storage modulus ( $E'$ ) at 120°C of not less than 10 MPa.

14. The substrate of claim 12, which is not crosslinked during or after a forming process thereof.

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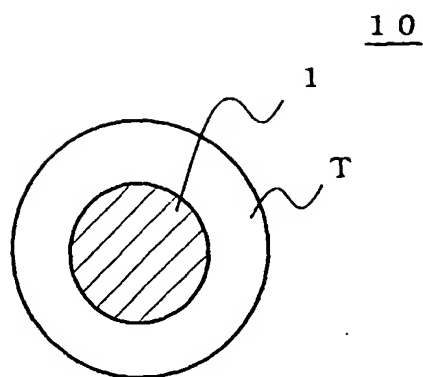
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**Fig. 1 (a)**



**Fig. 1 (b)**

